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Utilization of Gambier (*Uncaria Gambier,* Roxb.) Combined with Albumin in Formation of Natural Rigid Foam

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The treatment in this study was a variation in the amount of Gambier extract added, namely 16 g, 20 g and 24 g in 89.5 g of a mixture that also contained 23 g of albumin powder, 60 g of water, 5 g of hexamine and 1.5 g of para-Toluene Sulfonic Acid. Observations for the quality of the foam formed were carried out for several parameters such as Foam density, compressive strength, degree of development, pH modification and porosity. Observation of foam pores was conducted with an optical microscope (Bock and Norris, 2019). As a result, the best treatment combination was 24g Gambier extract per 89.5g of mixture prepared with pan-dried albumin. Microscopy observation showed that pan dried albumin powder resulted in 55% -65% pores and foam dried albumin powder pores were 41% - 50%. This study has shown the potential for utilizing Gambier extract in producing derivative products by combining with albumin. These results will be a reference for the development of other derivative products.

Keywords: Gambier extract, albumin powder, foam dried, pan dried, rigid foam, closed cells.

INTRODUCTION

Gambier (*Uncaria Gambier, Roxb*) is a plant belonging to the Rubiaceae family and in Indonesia is the world's main exporter and produced of Gambier and most of it comes from West Sumatra (Kasim *et al*., 2015; Hosen, 2017). This plant produces a lot of bioactive components with the main components being catechins and tannins (Anggraini *et al*, 2021) and contains ingredients insoluble alcohol about 34%, insoluble material 33% water, and 15% water content (Lukas *et al*., 2019). Gambier is one of the commodities of industrial plants which has high economic value as well as a good prospect for farmers and suppliers of foreign countries (Rauf *et al*., 2015).

The main properties of tannins depend on the phenolic group contained in the tannin, and these properties are generally chemical and physical properties. The chemical properties of tannins have general properties, namely having a phenol group and being colloidal, therefore in the air they are colloidal and weakly acidic. All types of tannins can dissolve in water. Their solubility is large, and will increase if dissolved in hot air. Tannins will dissolve in organic solvents such as methanol, ethanol, acetone and other organic solvents. The physical properties of tannins are generally tannins have a high molecular weight and tend to be easily oxidized into a polymer, most tannins are amorphous and do not have a melting point (Nofita and Dewangga, 2021).

Figure 1. Flower bud of Gambier plants.

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Figure 2. Gambier processed product from Gambier plant.

Gambier is the market name commonly known as the result of extraction from the leaves of the Gambier plant (Rauf *et al.*, 2015). Gambier contains several chemical components, one of which is tannin. Tannins are defined as water-soluble phenolic compounds that are found in fruits, woods, seeds, and in many vegetable oils (Varilla *et al*., 2020). The extraction process in Gambier can be done to get tannin. This process separates the material from the mixture using a solvent suitable for the material. This process is stopped when there is an equilibrium between the solvent and the concentration in the plant cell. One way to do this is by screening to separate samples (Mukhriani, 2014).

Tannins can be classified based on their chemical structure into two main conventional groups, namely hydrolyzable and condensed tannins (Bacelo *et al*., 2016). Due to the enormous structural diversity of the tannins a systematic classification system based on specific structural characteristics and chemical properties would provide a convenient framework for further study. The observation that many tannins can be fractionated hydrolytically into their components, for example by treatment with hot water or with tannases, led to the classification of such tannins as hydrolysable tannins (Khanbabaee and Ree, 2002). Non hydrolysable oligomeric and poly meric proanthocyanidins were classified as condensed tannins. Therefore, the term hydrolysable tannins include both the gallotannins and the ellagitannins. Figure 1 schematically depicts the di erent tannin classes as well as their chemical reactivity.

One affordable natural ingredient for this is the Gambier extract that is produced in Lima Puluh Kota, Indonesia as it has a high tannin content > 20% at 64.90%. Gambier starts from extracting hot water from the leaves and branches of the Gambier plant (*Uncaria gambier,* Roxb.) Which is then deposited, drained, opened and dried. Tannins can be used as making insulating foam (Lagel *et al*., 2014). In recent years, tannin-based foam has been obtained from 95% natural substrates resulting in an environmentally friendly product. Tannins can also be used as renewable and biobased oligomers to produce foam and could provide a safer alternative, being more environmentally friendly, and bringing new functional properties such as low flammability and self-extinguishing (Lacoste *et al*., 2015).

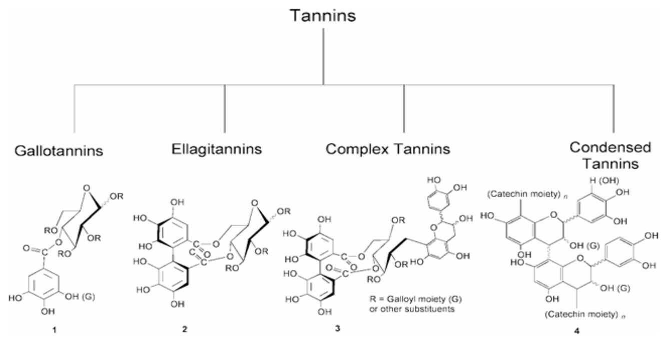

Figure 3. Systematic classification of tannins based on their structure and chemical properties (Khanbabaee, *et al*., 2002).

Foam based tannin can be made by mixing tannin, diethyl ether (which functions as a blowing agent), pentane (Li *et al*., 2013), but research was also conducted by (Szczurek *et al*., 2014) that informed, in the manufacture of liquid foam, using the formulation of hexamethylenetetramine (hexamin) as a hardener and para toluene sulfonid acid (pTSA) as a catalyst in order to make the foam produced more stable (Anova *et al*., 2018).

Tannins can also be used to produce foam and could provide a safer alternative, being more environmentally friendly and flame retardant. Lacoste *et al.* (2015) has investigated the manufacture of tannin-based foams from Quebracho wood, Pinus (Pinus radiate), Maritime Pine (Pinus pinaster) and Cemara bark (Picea abies) combined with protein from albumin to provide protein which the tannin can bind strongly to protein and can precipitate protein so as to maintain the porosity and texture of the foam.

This binding ability is due to the organic structure of proteins which contains N which forms strong complex compounds with carboxyl and hydroxyl groups in the tannins (Lacoste *et al*., 2015). Tannin content must be >20% for making foam and the higher the tannin content in a material, the stronger the bond between tannin and albumin (Deaville *et al*., 2010). This suggests that a tannin-rich material could be combined with duck egg albumin to make a rigid foam. One affordable natural ingredient for this is the Gambier extract that is produced in Lima Puluh Kota, Indonesia as it has a high tannin content >20% at 64.90% b/v. Rigid foams from tannin and albumin can be made because the chemical properties of tannins allow their polymerization and reactivity with albumin, which acts as a foaming agent and stabilizer. This combination produces a strong, lightweight, and environmentally friendly material with a wide range of potential applications.

The purpose of this study was to determine the effect of different concentrations of Gambier extract with two kinds of albumin powder in making foams to discover which combination could produce closed-cell foam (rigid foam) with the best characteristics, and the final application of foam can be used as insulation materials. Besides that, the use of tannins derived from Gambier extract in foam making can also support the use of Gambier in other forms as well as, and the foam produced is also made from natural ingredients which are cheaper and more affordable. Compared to foam that is already circulating on the market, e.g. polyurethane foam, natural rigid foam is safer because it doesn't use dangerous blowing agents such as hydrochlorofluorocarbon (HCFC), clorofluorocarbon (CFC) which can cause health problems and environmental damage.

The foam formed can be an environmentally friendly comparative product compared to poly urethane foam.Commercial. The foam from Polyurethane or PUR is produced from polyol, a synthetic petroleum derivative and generally uses blowing agents such as hydrochlorofluorocarbons (HCFC), clorofluorocarbons (CFCs) and volatile organic compounds such as methylene chloride which can cause health problems and environmental damage.

MATERIALS AND METHODS

***Materials*:** Gambier from Lima Puluh Kota Indonesia and duck egg albumin (Tegal duck / *Anasjavanica*) to be used as albumin powder, were used along with para-Toluene Sulfonic Acid (pTSA) as a catalyst and hexamine as a hardener obtained from PT. Sari Kimia-Padang.

***Equipment*:** The equipment used included a magnetic stirrer (AREC VelpScientifica-Europe, ultrasonic bath (Elma S300H), vacuum set evaporator (Buchi R-215), Germany, mixer (Philips), compressive strength test equipment (Cesare GaldabiniGallaret-Italy), pH meter (Delta OHM HD 2105.2, Italy), Olympus Light Microscope, ovens (Philip Harris Ltd), analytical scales (Kern ABJ 220-4M, Germany).

***Research design*:** This study uses a Factorial Complete Randomized Design (RAL Factorial) to show the effect of different amounts of Gambier extract and two kinds of albumin powder and the interaction of the relationship between these factors.

The first factor (A) was the amount of Gambier extract additions: with 16 g (A1), 20 g (A2), and 24 g (A3) in 89.5 g of the mixture which also contained 23 g albumin powder, 60 g water, 5 g hexamine and 1.5 g pTSA. Factor B was albumin powder which was prepared two different ways: pan dried (B1) and foaming dried (B2). The purpose of using 2 (two) drying methods is to see the difference in the result of the drying technique in obtaining of albumin powder against the resulting of rigid foam.

Table 1. Codes used for different Gambier extract amounts and albumin powder types.

Albumin drying method	Gambier extract (g) / 89.5 g mix		
	16 (A1)	20 (A2)	24 (A3)
Pan dried (B1)	A1B1	A2B1	A3B1
Foam dried (B2)	A1B2	A2B2	A3B2

*Results of each parameter were analysed using the F test and any statistically significant differences further tested using *Duncan's New Multiple Range Test* (DNMRT) at the 5% level

***Preparation of Gambier extract*:** Gambier was mashed until it passed an 80 mesh sieve, put into 250 ml conical flask, then distilled water, added using a 1:10 ratio of Gambier extract to distilled water. This was placed in an ultrasonic bath for ± 1 hour, then filtered. The filtrate was then evaporated using a water bath at ± 60ºC until a dried Gambier extract was obtained (Rini *et al*., 2021).

Preparation of albumin powder with thin layer drying (pan drying) and foaming drying methods

***Pan drying*:** Egg whites were separated from yolks then put into a baking dish measuring 25.5 cm x 25.5 cm and put into a 85ºC oven for ± 26 hours – the resulting material was passed through a 80 mesh filter.

***Foaming drying*:** Egg whites were separated from yolks then beaten using a mixer (± 20 minutes, 900 rpm), then put in a baking dish measuring 25.5 cm x 25.5 cm and dried in an 85ºC oven for ± 12 hours. The resulting material was passed through an 80 mesh filter.

***Making rigid foam*:** 23 g albumin powder was mixed with 60 g water then beaten ± 10 minutes until homogeneous using a speed of 1400 rpm. The appropriate amount of gambier extract was added along with 5 g of hexamine and 1.5 g of pTSA. The mixture was stirred until homogeneous, then put into an 85ºC oven for ± 2 hours.

***Observations*:** Foam density (Szczurek *et al*., 2014), compressive strength (Lacoste *et al*., 2015), degree of development (Rohaeti and Suyanta, 2011), pH modification (Lacoste *et al*., 2015; Syukri *et al*., 2013; Syukri *et al*,, 2024) and porosity (Lacoste *et al*., 2015) were measured. Observation of foam pores was conducted with an optical microscope (Bock and Norris, 2019).

RESULTS AND DISCUSSION

The mechanism of tannin albumin foam formation is the forming of bonds in tannin and albumin molecules while stirring rapidly so that reactions occurring between tannin hydroxyl groups and proteins from albumin trap air increasing the volume (Pizzi, 2019).

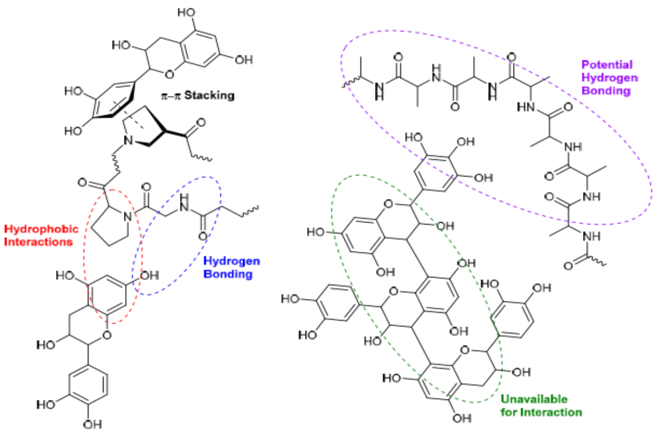

Figure 4. The possible hydrophobic interaction and hydrogen bondings between flavanol monomer or tannins with a protein (Watrelot and Norton, 2020).

Besides that, for rigid foams the primary and secondary hydroxyl group terminated polyether polyols are most important, followed by polyester polyols which have the oldest application.

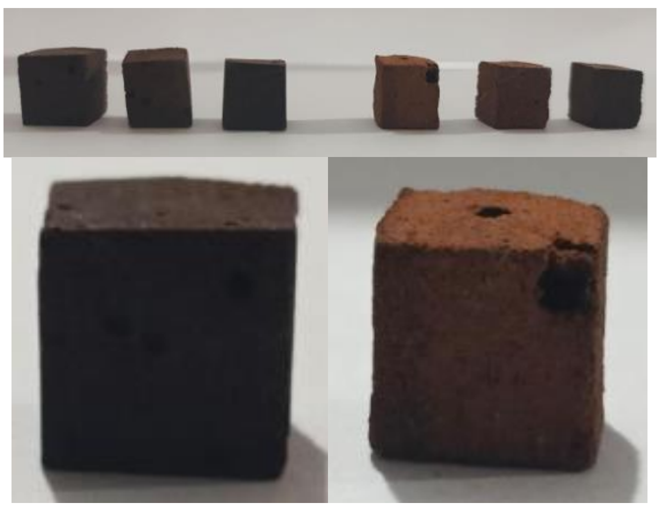

Figure 5. The appearance of the rigid foam from tannin and albumin.

***Foam density* (*Table 2, 3*):** Density is one indicator of mechanical strength (Firdaus, 2014), albumin powder prepared by pan drying resulted in lower density foam. The lowest foam density values were obtained with the A1B1 and A2B1 treatments (0.07 g / cm3) and the highest in the A1B2 and A3B2 treatments (0.27 g / cm3). Both the concentration of Gambier extract and the albumin drying method significantly affected (P <0.05) the foam density value. According to Lacoste *et al* (2015), rigid foam must have a density value of <0.3 g / cm3. So foams resulting from all concentrations of extract with this protein were low enough to be classified as rigid foam.

The higher the value of foam density, the higher the thermal conductivity (Lacoste *et al*., 2015). Anova *et al.* (2018) produced Gambier/albumin foam with density values ranging from 0.27 g / cm3-0.29 g / cm3. This type of foam can be used for decorative panels or internal and external insulation materials (Szczurek *et al*., 2014).

***Compressive strength (Table 2, 3)*:** Compressive strength is the ability of a material to maintain its integrity under pressure and indicates how much pressure can be received by the material (material) before it breaks so that it cannot be used again as intended (Abdulgani, 2014). Generally, for foam, the higher the foam density value, the higher the compressive strength.

The compressive strength of the foam was found to increase with the amount of Gambier extract. This was to be expected as the higher the amount of tannin extract, the greater the number of complex bonds between tannin and albumin (Lacoste *et al*., 2015). The lowest compressive strength value was from the A2B2 treatment which was 1.11kg / cm2 and the highest was obtained in the A3B1 treatment which was 8.63 kg / cm^2 more than twice the value of commercially available PUR and PIR foams. It was found that there was a statistically significant interaction (P <0.05) between the Gambier extract concentration with the method of making albumin powder on the compressive strength of the foam. According to SNI 3638 (2012), the compressive strength value of rigid foam must be below 10.08 kg / cm^2.

***Swelling degree (Table 2, 3)*:** The swelling degree determines the capacity of a material to absorb liquid until equilibrium occurs. The swelling degree varied from 24% -75%. The highest swelling degree was obtained in the A1B1 treatment which was 75% and the lowest was the A2B2 treatment which was 24.00%.

Link *et al.* (2011) state that for rigid foam the swelling degree is in the range of 10%-60% while Anova *et al.* (2018) found the lowest swelling degree was 55 %.

This present study suggests that foam dried albumin powder and the highest concentration of Gambier extract (A3B2) gives the best swelling degree at 43% The addition of Gambier extract and albumin had a significant effect and there was a statistically significant interaction between these two factors (P <0.05) on the swelling degree.

Rohaeti and Suyanta (2011) state that the swelling degree is an indication of the amount of cross-linking between the constituent materials in foam synthesis. The greater the swelling degree the greater the number of cross bonds in the structure and water used as a solvent can penetrate the foam network while a low swelling degree indicates a dense foam which is hard for water molecules to penetrate.

***pH (Table 2, 3)*:** The pH value of the foam obtained ranged from 7.17-7.53. The lowest pH was A3B1 treatment (7.17) and the highest A3B2 (7.53). According to Lacoste *et al.*

Table 2. Summary of results for foam made from pan-dried albumin powder.

Ttreatment	Parameter				
	Density (g/cm^3)	Compressive strength (kg/cm^2)	Swelling degree (%)	pH	Porosity (%)
A1B1	0.07±0.03a	5.40±0.59b	75.65±6.94b	7.36±0.04a	82.03±7.70b
A2B1	0.07±0.02a	6.70±0.24b	66.95±2.41b	7.23±0.01a	82.69±4.78b
A3B1	0.23±0.02a	8.63±0.10b	41.95±4.02b	7.17±0.02a	45.08±4.25b

Note: values accompanied by the same upper case letters indicate no significant difference between the different concentrations of extract the 5% level. Values with the same lower case letter are not statistically different to the equivalent extract concentration foam made with Foam-dried albumin at the 5% level. Errors are expressed as ± one standard deviation

Table 3. Summary of results for foam made from foam-dried albumin powder.

Treatment	Parameter				
	Density (g/cm^3)	Compressive strength (kg/cm^2)	Swelling degree (%)	pH	Porosity (%)
A1B2	0.27±0.02b	1.47±0.09a	33.48±3.65a	7.31±0.01b	33.92±4.12a
A2B2	0.22±0.00b	1.11±0.14a	24.00±1.73a	7.45±0.03b	47.73±0.47a
A3B2	0.27±0.02b	2.19±0.05a	43.13±2.13a	7.53±0.03b	35.20±5.65a

Note: Values accompanied by the same upper case letters indicate no significant difference between the different concentrations of extract the 5% level. Values with the same lower case letter are not statistically different to the equivalent extract concentration foam made with Pan-dried albumin at the 5% level. Errors are expressed as ± one standard deviation.

(2015) that the pH value of tannin and albumin-based foam is 7.1.

The addition of Gambier extract and the method of making albumin powder significantly affected the pH value of the foam produced and there was a significant interaction ($P < 0.05$) between the concentration of Gambier powder and the method of making albumin powder on the pH value. The best pH value for foam was made from pan-dried albumin. Lacoste et al (2015) also found the using pan-dried albumin powder resulted in a foam with a pH value that was not too high.

Acidic or basic conditions accelerate the condensation reaction between amine and amide groups and influence the position of the covalent bonds formed during manufacture, Increasing or decreasing the pH value will also affect the reactivity of the aldehydes in tannins (Lacoste *et al*., 2015).

***Porosity (Table 2, 3)*:** The highest porosity foams were A2B1 (82.69%) and A2B2 (47.73%) while the lowest were A3B1 (45.08%) and A1B2 (33.92%). Lacoste et al (2015) state that rigid foam should have a porosity value less than 68%, which was true of four of the samples A3B1, A1B2, A2B2, and A3B2.

The concentration of Gambier extract and albumin powder significantly affected ($P < 0.05$) the resulting porosity value and there was a statistically significant ($P < 0.05$) interaction between the concentration of Gambier powder and the albumin powder drying method on the porosity of the resulting rigid foam.

Changes to the concentration of Gambier extract used could be expected to change the resulting porosity value and cell size, as does stirring speed, stirring time, surfactant addition and cooking time (Szczurek *et al*., 2014). The porosity of foam determines the microscopic structure of the foam produced and is closely related to the density and compressive strength. Porosity is a property that tells us about the volume and quantity of pores with certain diameter. A high porosity value reduces the compressive strength and can also be caused by the evaporation of water in the foam. A summary table of all the observations is shown in Table 2 and Table 3.

***Microscopy of foam*:** This was conducted by cutting the foam into thin slices using a microtom tool (Yeung *et al*., 2015) The pore structure was observed under a microscope at 100 x. Observation of the microscopic structure of the foam was carried out using a microtome. The sample that had been cut was placed on an objective glass and dripped with distillated water. Sample is then placed on the microscope and clamped with a clamp, the adjust the focus to clarify the image of the object and view it from the eyepiece. Images can be seen in Figures 6 and figures 7.

Appearance varied from a few large pores to smaller denser pores. The results were similar to Lacoste *et al.* (2015) who found that addition of an acid catalyst resulted in a brown foam with a closed-cell structure. As expected, concentration of Gambier extract and albumin powder type influenced density, porosity and microscopic appearance and cell size (Szczurek *et al*., 2014).

Porosity is closely related to foam structure; smaller denser pores result in lower porosity. High porosity value results from larger and more pores. Pan-dried albumin produced foam with larger pores with an average pore size ranging from 55%-65% of that of flexible foam while the foam obtained with foam-dried albumin powder had fewer smaller pores in a denser structure with an average pore size ranging from 41% -50% of flexible foam. For pan-dried albumin foams the higher concentration of extract resulted in the best pore

appearance, while low concentration Gambier extract formed the best foam structure from the foam-dried albumin.

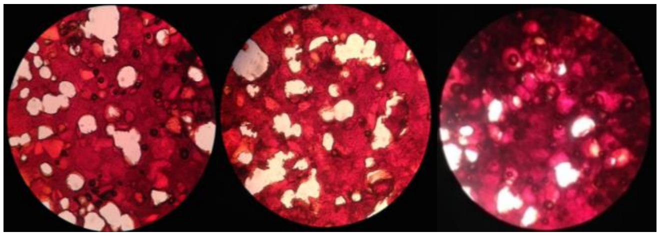

Figure 6. Appearance of foam pores structure (a) 16g Gambier extract and pan-dried albumin (A1B1), (b) 20g Gambier extract and pan-dried albumin (A2B1), (c) 24g Gambier extract and pan-dried albumin (A3B1).

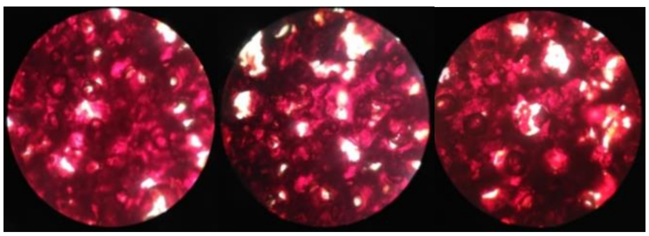

Figure 7. Appearance of foam pores structure (a) 16g Gambier extract and foam-dried albumin (A1B2), (b) 20g Gambier extract and foam-dried albumin (A2B2), (c) 24g Gambier extract and foam-dried albumin (A3B2).

From figures 2 and figures 3, porosity values it can be seen that the treatments resulting in the densest pore structure are A3B1 and A1B2. The pores in the Gambier/albumin foam were randomly shaped, small and broken up. This shows that the higher the addition of Gambier extract. The stiffer the quality of resulting foam.

Rigid foam from tannin and albumin can be used as an insulating material for the interior and exterior of wooden doors, heavy absorbent for metal, construction panels and packaging for soft goods and its tend to be more biodegradable than synthetic materials such as petroleum-based polyurethane. After use, these materials can be broken down into organic compounds that do not pollute the environment.

***Conclusion*:** In this study, foam has been developed using Gambier extract combined with albumin. The combination of 24 g of Gambier extract per 89.5 g of mixture produces a good foam product so that this formula can be further developed. In addition, the manufacture of effective dry albumin can be done by the pan-dried method. The information obtained from this study can be a reference for the development of Gambier-based products in the future. The development of Gambier-based products is expected to increase the diversification of Gambier derivative products so that farmers will be able to develop further.

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***SDGs addressed*:** Industry, Innovation, and Infrastructure, Responsible Consumption and Production, Climate Action.

***Policy referred*:** Green Innovation and Sustainable Development Policies; Waste Valorization and Natural Resource Utilization Policies; Climate Action and Pollution Reduction Policy Goals.

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